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A Multislice Theory of Electron Inelastic Scattering in a Solid

BY Z. L. WANG

Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, England

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Abstract

A multislice theory is proposed to solve Yoshioka's coupling equations for elastic and inelastic scattered high-energy electrons in a solid. This method is capable, in principle, of including the non-periodic crystal structures and the electron multiple scattering among all the excited states in the calculations. It is proved that the proposed theory for calculating the energy-filtered inelastic images [Wang (1989). *Acta Cryst.* A**45**, 193-199], based on the physical optics approach, is equivalent to the quantum-mechanical theory under some approximations. The basic theory of simulating the energy-filtered inelastic image of core-shell losses and thermal diffuse scattering is outlined.

1. Introduction

Electron inelastic scattering, as a general phenomenon in high-energy electron diffraction, has been widely used in investigating the structures of materials. The early theory of simulating inelastic waves was due to Yoshioka (1957). Based on quantum-mechanical theory, he derived a set of coupled Schrödinger equations by considering the transitions between the ground state and the excited states of the crystal. Because of the complicated coupling between those equations, a proper method has not been established to solve all the inelastic components. Some work has been done to consider the effect of inelastic scattering on elastic scattering in a first-order approximation. Besides the crystal potential due to the atomic arrangement, Yoshioka (1957) predicted that there will be a real addition to the crystal potential from virtual inelastic scattering as well as the imaginary absorption correction. The corrections due to single-electron inelastic scattering were calculated using the Thomas-Fermi atomic model (Yoshioka, 1957) and atomic wave functions (Whelan, 1965*a*). This correction has also been calculated by Humphreys & Hirsch (1968) for different inelastic excitation processes. The contribution due to plasmon scattering was studied by Radi (1970) and Yoshioka & Kainuma (1962).

All the above work was carried out for the purpose of considering the effect of inelastic scattering on electron elastic scattering. An attempt to solve the inelastic waves from Yoshioka's coupling equations was made by Howie (1963). Based on the Bloch-wave approach under small-angle approximation, he gave some analytical solutions for phonon excitations under some simplified conditions. By neglecting the joint transitions among the excited states, this Bloch-wave approach was extensively developed by Rez, Humphreys & Whelan (1977), and Rez (1983). Serneels, Haentjens & Gevers (1980) have proposed an iteration method for solving Yoshioka's coupling equations, which assumes a periodic structure of the crystal. So far, there is no method available which can solve Yoshioka's coupling equations for a general solid structure considering all possible transitions.

The multislice theory, developed by Cowley & Moodie (1957), has been successfully applied in image simulation for high-energy electrons in crystals with defects. It has been shown that this approach is equivalent to the quantum-mechanical theory (Goodman & Moodie, 1974; Ishizuka & Uyeda, 1977). In other words, the multislice theory is a method of solving the Schrödinger equation for high-energy electrons under some approximations. In this paper, we start from the integral forms of Yoshioka's coupling equations to derive their solutions in the scheme of multislice theory. The final solutions contain the multiple elastic and inelastic excitations in a solid. Some solutions of the first-order approximation will be examined and compared with the early theory developed for calculating the energy-filtered inelastic images in electron microscopy (Wang, 1989*a*). The detailed theories of calculating energy-filtered inelastic core-shell images and thermal diffuse scattering will be outlined.

2. The general solution of Yoshioka's coupling equations using the multislice method

First, one follows Yoshioka's method for deriving the coupling equations. Consider the interaction of an incident electron with a solid. The Schrödinger equation of the system is

$$\left(-\frac{\hbar^2}{2m_0}\nabla^2 + H_c + H'\right)\Phi = E\Phi, \qquad (1)$$

where $-(\hbar^2/2m_0)\nabla^2$ is the kinetic energy of the electron, H_c is the crystal Hamiltonian and H' describes the interaction between the electron and the solid. $\Phi(\mathbf{r}, \mathbf{r}_1, \ldots, \mathbf{r}_M)$ is the wave function of the system,

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which depends on $\mathbf{r}$, the coordinates of the incident electron, and on $\mathbf{r}_1, \ldots, \mathbf{r}_M$, the coordinates of the electrons and ions of the crystal. Neglecting exchange effects one can write

$$\Phi(\mathbf{r}, \mathbf{r}_1, \ldots, \mathbf{r}_M) = \sum_n a_n(\mathbf{r}_1, \ldots, \mathbf{r}_M)\Psi_n(\mathbf{r}), \quad (2)$$

where a_n is the wave function of the crystal in its nth excited state of energy ε_n so that

$$H_c a_n = \varepsilon_n a_n. \quad (3)$$

Ψ_0 in (2) describes the elastic scattered wave of energy $E_0 = E$, and Ψ_n describes the inelastically scattered wave of energy $E_n = E - \varepsilon_n$, with $n = 1, 2, \ldots, m$. Substituting (2) and (3) into (1), multiplying by a_n^*, and integrating over the coordinates $\mathbf{r}_1, \ldots, \mathbf{r}_M$, one obtains

$$(\nabla^2 + k_0^2)\Psi_0 = \sum_m (2m_0/\hbar^2)H'_{0m}(\mathbf{r})\Psi_m, \quad (4a)$$

$$(\nabla^2 + k_n^2)\Psi_n = \sum_m (2m_0/\hbar^2)H'_{nm}(\mathbf{r})\Psi_m, \quad (4b)$$

where

$$k_n^2 = (2m_0/\hbar^2)E_n \quad (4c)$$

and

$$H'_{nm} = \int a_n^* H' a_m \, d\mathbf{r}_1, \ldots, d\mathbf{r}_M. \quad (4d)$$

These are Yoshioka's coupling equations for inelastic scattering. Our goal is to find the multislice solution of these equations. For convenience of calculation, we write (4a) and (4b) in matrix form,

$$\nabla^2 \begin{bmatrix} \Psi_0 \\ \Psi_1 \\ \vdots \\ \Psi_m \end{bmatrix} + \begin{bmatrix} k_0^2\Psi_0 \\ k_1^2\Psi_1 \\ \vdots \\ k_m^2\Psi_m \end{bmatrix} = \frac{2m_0}{\hbar^2} \begin{bmatrix} H'_{00} & H'_{01} & \ldots & H'_{0m} \\ H'_{10} & H'_{11} & \ldots & H'_{1m} \\ \vdots & \vdots & \vdots & \vdots \\ H'_{m0} & H'_{m1} & \ldots & H'_{mm} \end{bmatrix} \begin{bmatrix} \Psi_0 \\ \Psi_1 \\ \vdots \\ \Psi_m \end{bmatrix}. \quad (5)$$

Equation (5) is an inhomogeneous equation. The H' matrix is an $m \times m$ transition matrix. By defining $\Psi_n = \exp(i\mathbf{k}_n \cdot \mathbf{r})\varphi_n$, using the Green function method and assuming $\varepsilon_n \ll E_0$, (5) can be written in the form of (6) (see Appendix A):

$$\begin{bmatrix} \varphi_0(\mathbf{r}) \\ \varphi_1(\mathbf{r}) \\ \vdots \\ \varphi_m(\mathbf{r}) \end{bmatrix} = I - \int \frac{2m_0}{4\pi\hbar^2} d\mathbf{r}' \times \begin{bmatrix} F(\mathbf{r}-\mathbf{r}', \mathbf{k}_0) & 0 & \ldots & 0 \\ 0 & F(\mathbf{r}-\mathbf{r}', \mathbf{k}_1) & \ldots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \ldots & F(\mathbf{r}-\mathbf{r}', \mathbf{k}_m) \end{bmatrix} \times \begin{bmatrix} H'_{00}(\mathbf{r}') & H'_{01}(\mathbf{r}') & \ldots & H'_{0m}(\mathbf{r}') \\ H'_{10}(\mathbf{r}') & H'_{11}(\mathbf{r}') & \ldots & H'_{1m}(\mathbf{r}') \\ \vdots & \vdots & \vdots & \vdots \\ H'_{m0}(\mathbf{r}') & H'_{m1}(\mathbf{r}') & \ldots & H'_{mm}(\mathbf{r}') \end{bmatrix} \begin{bmatrix} \varphi_0(\mathbf{r}') \\ \varphi_1(\mathbf{r}') \\ \vdots \\ \varphi_m(\mathbf{r}') \end{bmatrix} \quad (6a)$$

where F is defined as

$$F(\mathbf{r}-\mathbf{r}', \mathbf{k}_n) = \frac{\exp\{i[k_n|\mathbf{r}-\mathbf{r}'| - \mathbf{k}_n \cdot (\mathbf{r}-\mathbf{r}')]\}}{|\mathbf{r}-\mathbf{r}'|} \quad (6b)$$

and I is a matrix defined as

$$I = \begin{bmatrix} 1 \\ 1 \\ \vdots \\ 1 \end{bmatrix}. \quad (6c)$$

This expression is valid when the high-energy electron scattering satisfies the following conditions:

(1) $\alpha^2 \ll 1$, α is the scattering angle.

(2) $|\mathbf{r}-\mathbf{r}'| \simeq z - z'$.

Thus

$$k_n|\mathbf{r}-\mathbf{r}'| - \mathbf{k}_n \cdot (\mathbf{r}-\mathbf{r}') \simeq k_n \frac{|\mathbf{b}-\mathbf{b}'|^2}{2(z-z')}.$$

Here $\mathbf{b} = (x, y)$ and $\mathbf{b}' = (x', y')$.

Following the method introduced by Ishizuka & Uyeda (1977), one finds that (6a) becomes

$$\begin{bmatrix} \varphi_0(\mathbf{b}, z) \\ \varphi_1(\mathbf{b}, z) \\ \vdots \\ \varphi_m(\mathbf{b}, z) \end{bmatrix} = I + \left(-\frac{i}{\hbar v}\right) \times \int_{z'=-\infty}^{z'=z} \int \begin{bmatrix} P_0(\mathbf{b}-\mathbf{b}', z-z') & 0 & \ldots & 0 \\ 0 & P_1(\mathbf{b}-\mathbf{b}', z-z') & \ldots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \ldots & P_m(\mathbf{b}-\mathbf{b}', z-z') \end{bmatrix} \times \begin{bmatrix} H'_{00}(\mathbf{b}', z') & H'_{01}(\mathbf{b}', z') & \ldots & H'_{0m}(\mathbf{b}', z') \\ H'_{10}(\mathbf{b}', z') & H'_{11}(\mathbf{b}', z') & \ldots & H'_{1m}(\mathbf{b}', z') \\ \vdots & \vdots & \vdots & \vdots \\ H'_{m0}(\mathbf{b}', z') & H'_{m1}(\mathbf{b}', z') & \ldots & H'_{mm}(\mathbf{b}', z') \end{bmatrix} \begin{bmatrix} \varphi_0(\mathbf{b}', z') \\ \varphi_1(\mathbf{b}', z') \\ \vdots \\ \varphi_m(\mathbf{b}', z') \end{bmatrix} d\mathbf{b}' \, dz', \quad (7a)$$

where P_n is defined as a propagation function,

$$P_n(\mathbf{b}, z) = (1/i\lambda z)\exp(ik_n b^2/2z). \quad (7b)$$

Here λ is the wavelength of the electron and v is its velocity.

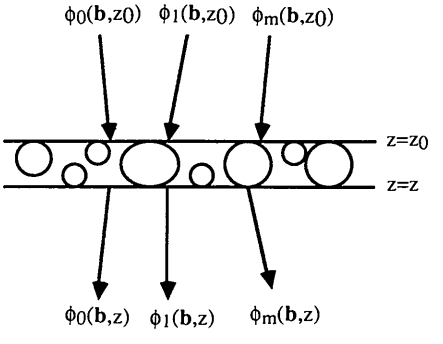

Fig. 1. A schematic diagram illustrating the waves before and after transmission through a crystal slice.

Equation (7a) is an integral equation. In order to find the multislice solution of this equation, we need to derive the relationship between the waves, $\varphi_n(\mathbf{b}, z_0)$, going into a crystal slice, with thickness $(z-z_0)$, and those, $\varphi_n(\mathbf{b}, z)$, after being scattered by the crystal slice, as shown in Fig. 1. Now let us consider the nth row of (7a):

$$\begin{aligned}\varphi_n(\mathbf{b}, z) &= 1+(-i/\hbar v)\iint_{z'=-\infty}^{z'=z} P_n(\mathbf{b}-\mathbf{b}', z-z')\\ &\quad\times\sum_m H'_{nm}(\mathbf{b}', z')\varphi_m(\mathbf{b}', z')\,\mathrm{d}\mathbf{b}'\,\mathrm{d}z'\\ &= 1+(-i/\hbar v)\iint_{z''=-\infty}^{z''=z_0} P_n(\mathbf{b}-\mathbf{b}'', z-z'')\\ &\quad\times\sum_m H'_{nm}(\mathbf{b}'', z'')\varphi_m(\mathbf{b}'', z'')\,\mathrm{d}\mathbf{b}''\,\mathrm{d}z''\\ &\quad+(-i/\hbar v)\iint_{z'=z_0}^{z'=z} P_n(\mathbf{b}-\mathbf{b}', z-z')\\ &\quad\times\sum_m H'_{nm}(\mathbf{b}', z')\varphi_m(\mathbf{b}', z')\,\mathrm{d}\mathbf{b}'\,\mathrm{d}z'. \qquad (8)\end{aligned}$$

We use the following properties (Ishizuka & Uyeda, 1977):

$$\int P_n(\mathbf{b}, z)\,\mathrm{d}\mathbf{b} = 1; \qquad (9a)$$

$$P_n(\mathbf{b}-\mathbf{b}', z-z') = \int_{\Sigma} P_n(\mathbf{b}-\mathbf{b}'', z-z'')P_n(\mathbf{b}''-\mathbf{b}, z''-z')\,\mathrm{d}\mathbf{b}''. \qquad (9b)$$

Σ is a plane positioned at z'' between z and z'. Then (8) can be rewritten as

$$\begin{aligned}\varphi_n(\mathbf{b}, z) &= \int[1+(-i/\hbar v)\iint_{z''=-\infty}^{z''=z_0} P_n(\mathbf{b}_0-\mathbf{b}'', z_0-z'')\\ &\quad\times\sum_m H'_{nm}(\mathbf{b}'', z'')\varphi_m(\mathbf{b}'', z'')\,\mathrm{d}\mathbf{b}''\,\mathrm{d}z''\\ &\quad\times P_n(\mathbf{b}-\mathbf{b}_0, z-z_0)]\,\mathrm{d}\mathbf{b}_0\\ &\quad+(-i/\hbar v)\iint_{z'=z_0}^{z'=z} P_n(\mathbf{b}-\mathbf{b}', z-z')\\ &\quad\times\sum_m H'_{mn}(\mathbf{b}', z')\varphi_m(\mathbf{b}', z')\,\mathrm{d}\mathbf{b}'\,\mathrm{d}z'. \qquad (10)\end{aligned}$$

Comparing the first term in (10) with (8) for $z=z_0$, we can write

$$\begin{aligned}\varphi_n(\mathbf{b}, z) &= \int P_n(\mathbf{b}-\mathbf{b}_0, z-z_0)\varphi_n(\mathbf{b}_0, z_0)\,\mathrm{d}\mathbf{b}_0\\ &\quad+(-i/\hbar v)\iint_{z'=z_0}^{z'=z} P_n(\mathbf{b}-\mathbf{b}', z-z')\\ &\quad\times\sum_m H'_{nm}(\mathbf{b}', z')\varphi_m(\mathbf{b}', z')\,\mathrm{d}\mathbf{b}'\,\mathrm{d}z'. \qquad (11)\end{aligned}$$

In order to solve (11), one expands φ_n in powers of $(-i/\hbar v)$ (Ishizuka & Uyeda, 1977):

$$\varphi_n(\mathbf{b}, z) = \sum_{L=0}^{\infty}(-i/\hbar v)^L f_L^{(n)}(\mathbf{b}, z). \qquad (12)$$

We put (12) into (11), and by equating the coefficients of $(-i/\hbar v)^L$, we get

$$f_0^{(n)}(\mathbf{b}, z) = \int P_n(\mathbf{b}-\mathbf{b}_0, z-z_0)\varphi_n(\mathbf{b}_0, z_0)\,\mathrm{d}\mathbf{b}_0 \qquad (13a)$$

$$f_L^{(n)}(\mathbf{b}, z) = \iint_{z'=z_0}^{z'=z} P_n(\mathbf{b}-\mathbf{b}', z-z')\times\sum_m H'_{nm}(\mathbf{b}', z')f_{L-1}^{(m)}(\mathbf{b}', z')\,\mathrm{d}\mathbf{b}'\,\mathrm{d}z'. \qquad (13b)$$

If H'_{nm} varies slowly in the region of $\Delta z = (z-z_0)$ and also $k_0 \simeq k_n$, then the solution of (13) can be proved as (see Appendix B)

$$\begin{bmatrix} f_L^{(0)}(\mathbf{b}, z)\\ f_L^{(1)}(\mathbf{b}, z)\\ \vdots\\ f_L^{(m)}(\mathbf{b}, z)\end{bmatrix} = \frac{1}{L!}\int \mathrm{d}\mathbf{b}_0 \begin{bmatrix} P_0(\mathbf{b}-\mathbf{b}_0, \Delta z) & 0 & \cdots & 0\\ 0 & P_1(\mathbf{b}-\mathbf{b}_0, \Delta z) & \cdots & 0\\ \vdots & \vdots & \vdots & \vdots\\ 0 & 0 & \cdots & P_m(\mathbf{b}-\mathbf{b}_0, \Delta z)\end{bmatrix} \times \begin{bmatrix} h'_{00}(\mathbf{b}_0, \Delta z) & h'_{01}(\mathbf{b}_0, \Delta z) & \cdots & h'_{0m}(\mathbf{b}_0, \Delta z)\\ h'_{10}(\mathbf{b}_0, \Delta z) & h'_{11}(\mathbf{b}_0, \Delta z) & \cdots & h'_{1m}(\mathbf{b}_0, \Delta z)\\ \vdots & \vdots & \vdots & \vdots\\ h'_{m0}(\mathbf{b}_0, \Delta z) & h'_{m1}(\mathbf{b}_0, \Delta z) & \cdots & h'_{mm}(\mathbf{b}_0, \Delta z)\end{bmatrix}^L \begin{bmatrix}\varphi_0(\mathbf{b}_0, z_0)\\ \varphi_1(\mathbf{b}_0, z_0)\\ \vdots\\ \varphi_m(\mathbf{b}_0, z_0)\end{bmatrix} \qquad (14)$$

where h' is defined as

$$h'_{nm} = \int_{z_0}^{z} H'_{nm}(\mathbf{b}, z')\,\mathrm{d}z'. \qquad (15a)$$

Equation (15a) can be written in the form of (15b) for $(z-z_0)\to 0$:

$$h'_{nm} \simeq H'_{nm}(\mathbf{b}, z)\,\Delta z. \qquad (15b)$$

Combining (14) and (12), one gets the result

$$\begin{bmatrix}\varphi_0(\mathbf{b}, z)\\ \varphi_1(\mathbf{b}, z)\\ \vdots\\ \varphi_m(\mathbf{b}, z)\end{bmatrix} = \int \mathrm{d}\mathbf{b}_0 \begin{bmatrix} P_0(\mathbf{b}-\mathbf{b}_0, \Delta z) & 0 & \cdots & 0\\ 0 & P_1(\mathbf{b}-\mathbf{b}_0, \Delta z) & \cdots & 0\\ \vdots & \vdots & \vdots & \vdots\\ 0 & 0 & \cdots & P_m(\mathbf{b}-\mathbf{b}_0, \Delta z)\end{bmatrix} \times \exp\left\{-\frac{i}{\hbar v}\begin{bmatrix} h'_{00}(\mathbf{b}_0, \Delta z) & h'_{01}(\mathbf{b}_0, \Delta z) & \cdots & h'_{0m}(\mathbf{b}_0, \Delta z)\\ h'_{10}(\mathbf{b}_0, \Delta z) & h'_{11}(\mathbf{b}_0, \Delta z) & \cdots & h'_{1m}(\mathbf{b}_0, \Delta z)\\ \vdots & \vdots & \vdots & \vdots\\ h'_{m0}(\mathbf{b}_0, \Delta z) & h'_{m1}(\mathbf{b}_0, \Delta z) & \cdots & h'_{mm}(\mathbf{b}_0, \Delta z)\end{bmatrix}\right\} \times \begin{bmatrix}\varphi_0(\mathbf{b}_0, z_0)\\ \varphi_1(\mathbf{b}_0, z_0)\\ \vdots\\ \varphi_m(\mathbf{b}_0, z_0)\end{bmatrix}. \qquad (16)$$

Equation (16) gives a general relationship between all the elastic and inelastic waves before and after penetrating a crystal slice. The exponential matrix indicates the multiple scattering of the electrons among all these states.

In order to get some simplified results, we consider the first-order approximation of (16). Assuming $|H_{nm}| \ll |H_{nn}|$ and $\sigma|H_{nm}| \ll 1$ for $n \neq m$, with $\sigma = 1/\hbar v$, then the exponential term of the matrix can be written as a summation of a diagonal matrix and a non-diagonal matrix:

$$\begin{aligned}\exp\{-i\sigma[\ldots]\} &= \exp\left\{-i\sigma\left(\begin{bmatrix} h'_{00} & 0 & \ldots & 0 \\ 0 & h'_{11} & \ldots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \ldots & h'_{mm} \end{bmatrix} + \begin{bmatrix} 0 & h'_{01} & \ldots & h'_{0m} \\ h'_{10} & 0 & \ldots & h'_{1m} \\ \vdots & \vdots & \vdots & \vdots \\ h'_{m0} & h'_{m1} & \ldots & 0 \end{bmatrix}\right)\right\} \\ &\simeq \exp\left\{-i\sigma\begin{bmatrix} h'_{00} & 0 & \ldots & 0 \\ 0 & h'_{11} & \ldots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \ldots & h'_{mm} \end{bmatrix}\right\} \\ &\quad \times \exp\left\{-i\sigma\begin{bmatrix} 0 & h'_{01} & \ldots & h'_{0m} \\ h'_{10} & 0 & \ldots & h'_{1m} \\ \vdots & \vdots & \vdots & \vdots \\ h'_{m0} & h'_{m1} & \ldots & 0 \end{bmatrix}\right\} \\ &\simeq \begin{bmatrix} \exp(-i\sigma h'_{00}) & -i\sigma h'_{01} & \ldots & -i\sigma h'_{0m} \\ \vdots & \vdots & \vdots & \vdots \\ -i\sigma h'_{m0} & -i\sigma h'_{m1} & \ldots & \exp(-i\sigma h'_{mm}) \end{bmatrix}. \end{aligned} \tag{17}$$

If we put (17) into (16), we have

$$\begin{bmatrix} \varphi_0(\mathbf{b}, z) \\ \varphi_1(\mathbf{b}, z) \\ \vdots \\ \varphi_m(\mathbf{b}, z) \end{bmatrix} = \begin{bmatrix} \{\exp(-i\sigma h'_{00})[\varphi_0(\mathbf{b}, z_0) - i\sigma \textstyle\sum_{n\neq 0} h'_{0n}\varphi_n(\mathbf{b}, z_0)]\} * P_0 \\ \{\exp(-i\sigma h'_{11})[\varphi_1(\mathbf{b}, z_0) - i\sigma \textstyle\sum_{n\neq 1} h'_{1n}\varphi_n(\mathbf{b}, z_0)]\} * P_1 \\ \vdots \\ \{\exp(-i\sigma h'_{mm})[\varphi_m(\mathbf{b}, z_0) - i\sigma \textstyle\sum_{n\neq m} h'_{mn}\varphi_n(\mathbf{b}, z_0)]\} * P_m \end{bmatrix} \tag{18}$$

where $*$ denotes the convolution of the propagation function with respect to $\mathbf{b}$.

For the elastic scattered wave, φ_0, the first term is the phase-grating result of the crystal slice, which is the elastic penetration of the incident elastic wave. The terms containing h'_{0n} are the 'transitions' of the electrons from the excited states to the ground state. For the excited states, φ_n, the first term is the elastic scattering of the incident inelastic wave in the slice; the h'_{mn} terms are the transitions from the other states, including the ground state, to the nth state. This term can be considered as the generation of the inelastic wave when the electron penetrates through the slice. Also it is obvious that (18) reduces to the multislice formula (Cowley & Moodie, 1957) of elastic scattering if all the inelastic transitions vanish, *i.e.* $h'_{mn} = 0$ for $n \neq m$.

If one knows the elastic incident wave before the electron strikes the crystal surface, then all the generated elastic and inelastic waves after the first slice can be calculated through (18). Then these waves can be taken as the incident waves for the second slice; the waves after penetrating the second slice can also be calculated through (18). So all the waves after penetrating through the crystal can be obtained.

3. Special cases of only one excited state

Let us consider the case of only one excited state. This case happens in most plasmon excitation and single-electron excitations. The crystal can be considered as either in its excited state or in its ground state. In practice, this corresponds to the case of the energy-filtered inelastic images from the electrons after exciting a plasmon loss or core-shell loss in electron microscopy. With $m = 1$, through some algebra, (16) reduces to the following forms:

$$\begin{aligned}\varphi_0(\mathbf{b}, z) = \{&\exp(-i\sigma h'_{00})[\cos(\sigma|h'_{10}|)\varphi_0(\mathbf{b}, z_0) \\ &- i\sin(\sigma|h'_{10}|)(h'_{01}/|h'_{01}|)\varphi_1(\mathbf{b}, z_0)]\} * P_0 \end{aligned} \tag{19a}$$

$$\begin{aligned}\varphi_1(\mathbf{b}, z) = \{&\exp(-i\sigma h'_{11})[\cos(\sigma|h'_{10}|)\varphi_1(\mathbf{b}, z_0) \\ &- i\sin(\sigma|h'_{10}|)(h'_{10}/|h'_{10}|)\varphi_0(\mathbf{b}, z_0)]\} * P_1. \end{aligned} \tag{19b}$$

3.1. *Case* 1: *energy-filtered inelastic image from a plasmon loss*

In (19), since $\cos(\sigma|h'_{10}|) \simeq 1 - (\sigma|h'_{10}|)^2/2 \simeq \exp[-(\sigma|h'_{10}|)^2/2]$ for $\sigma|h'_{10}| \ll 1$, we can define an absorption function μ as given by (20*a*) and an electron inelastic mean free path Λ as given by (20*b*):

$$\mu(\mathbf{b}, z_0) = (\sigma|h'_{10}|)^2/\Delta z, \tag{20a}$$

$$\Lambda(\mathbf{b}, z_0) = 1/\mu. \tag{20b}$$

After neglecting the transition of the electron from its excited state to the ground state, we can write (19) as

$$\begin{aligned}\varphi_0(\mathbf{b}, z) = \{&\exp(-i\sigma h'_{00}) \\ &\times[\exp(-\mu\Delta z/2)\varphi_0(\mathbf{b}, z_0)]\} * P_0 \end{aligned} \tag{21a}$$

$$\begin{aligned}\varphi_1(\mathbf{b}, z) = \{&\exp(-i\sigma h'_{11})[\exp(-\mu\Delta z/2)\varphi_1(\mathbf{b}, z_0) \\ &- i\sigma h'_{10}\varphi_0(\mathbf{b}, z_0)]\} * P_1. \end{aligned} \tag{21b}$$

In (21*a*), the $\exp(-\mu\Delta z/2)$ term describes the absorption effect of inelastic scattering. For plasmon excitations, h'_{01} may be taken as a product of a complex constant with a real function, and also $h'_{00} \simeq h'_{11}$; then (21) can be written in the form of (22) after a proper choice of the constant phase factor of the wave function φ_0:

$$\varphi_0(\mathbf{b}, z) = \{q(\mathbf{b})[\exp(-\mu\Delta z/2)\varphi_0(\mathbf{b}, z_0)]\} * P_0 \tag{22a}$$

$$\begin{aligned}\varphi_1(\mathbf{b}, z) = \{&q(\mathbf{b})[\exp(-\mu\Delta z/2)\varphi_1(\mathbf{b}, z_0) \\ &+ (\Delta z/\Lambda)^{1/2}\varphi_0(\mathbf{b}, z_0)]\} * P_1, \end{aligned} \tag{22b}$$

where $q(\mathbf{b})$ is defined as the slice transmission function,

$$q(\mathbf{b}) = \exp(-i\sigma h'_{00}). \tag{22c}$$

Equation (19) is the form of the theory proposed by Wang (1989*a*) for calculating energy-filtered inelastic images based on the semi-classical approach. In (22), the term containing μ indicates the absorption of inelastic excitation. The term $\Delta z/\Lambda$ is the probability of generating the plasmon loss while the electron is traveling through the slice. This means that the electron energy loss is a mean process depending on its average traveling distance. This is the classical concept of electron energy loss, which is still valid in quantum mechanics under the first-order approximation (Ritchie & Howie, 1988). Equation (22) can also be applied for single-electron excitations. Since (22) was derived directly from quantum mechanics, this inversely proves the equivalence of the previous theory (Wang, 1989*a*) with quantum mechanics under some approximation. The detailed calculations of the energy-filtered inelastic images of plasmon losses have been given elsewhere (Wang, 1989*a*, *b*).

3.2. *Case* 2: *total elastic and inelastic scattering*

The elastic and inelastic scattered waves, in principle, should be treated incoherently. In practice, however, if the electron energy loss is small, the inelastic process can be localized in a wide range along the beam direction. Also, no large momentum transfer effect is introduced in the direction perpendicular to the beam. Then it is valid to assume that the inelastic process may take place anywhere and one can assume that it takes place after transmission through the crystal. Hence the amplitudes of all components may be added together inside the crystal. The effect of electron energy loss is to perturb the wavelength. This was the semi-classical approach proposed for including the plasmon diffuse scattering in dynamical calculations (Wang, 1989*b*; Wang & Lu, 1988). The correspondence of this treatment could be derived from (19). If h'_{10} is a real function, then by adding the φ_0 and φ_1, one gets

$$[\varphi_0(\mathbf{b}, z)+\varphi_1(\mathbf{b}, z)] \simeq (\exp\{-i\sigma(h'_{00}+|h'_{10}|) \times [\varphi_0(\mathbf{b}, z_0)+\varphi_1(\mathbf{b}, z_0)]\}) * P_0. \quad (23)$$

This equation shows that the total scattered wave can be calculated approximately by assuming a real correction to the crystal potential. Actually this real correction, based on classical energy-loss theory, indicates the perturbation of electron energy loss to the wavelength depending on the scattering trajectory (Wang, 1989*b*).

3.3. *Single-electron core–shell excitations*

In electron diffraction, one inelastic process is the single-electron excitation, during which an electron is excited from one bound state to another state, such as the transition of a 1*s* electron to an ionization state, which is called the *K*-edge loss in electron energy-loss spectroscopy. Equation (21) can be applied to calculate the inelastic wave of single-electron core–shell excitations. In this section, one uses the atomic tight-binding approximation for deriving the transition matrix of this process.

The transition matrix of an inelastic transition from an initial ground state to the *n*th excited state can be written as (Howie, 1963)

$$H'_{n0}(\mathbf{r}, \mathbf{q}) = \exp(-i\mathbf{q}_{n0} \cdot \mathbf{r}) \sum_{\mathbf{g}} H_g^{n0} \exp(i\mathbf{g} \cdot \mathbf{r}), \quad (24)$$

where $\mathbf{q}$ is the wave vector of the excitation created in the lattice (*e.g.* exciton, phonon, plasmon *etc.*). $\mathbf{g}$ is the reciprocal-lattice vector. For single-electron excitation, it is possible to use the tight-binding approximation for calculating H_g^{nm} (Whelan, 1965*a*). Following the mathematical procedures given by Landau & Lifshitz (1977), one gets

$$H_g^{nm} = (4\pi e^2/V)[f_{nm}(\mathbf{g}-\mathbf{q})/|\mathbf{g}-\mathbf{q}|^2], \quad (25a)$$

where

$$f_{nm}(\mathbf{K}) = \langle n|\exp(-i\mathbf{K} \cdot \mathbf{r})|m\rangle, \quad (25b)$$

and $|n\rangle$ and $|m\rangle$ are the normalized one-electron atomic wave functions. V is the volume of the atom. We consider the contributions of all the atoms in the unit cell located at $\mathbf{R}_j$ ($j=1, 2, \ldots$), to the matrix element at position $\mathbf{r}$, and make use of (24) and (25); then

$$H'_{nm}(\mathbf{r}, \mathbf{q}) = (4\pi e^2/V_c) \sum_{\mathbf{g}} \sum_j [f^j_{nm}(\mathbf{g}-\mathbf{q})/|\mathbf{g}-\mathbf{q}|^2 \times \exp[i(\mathbf{g}-\mathbf{q}) \cdot (\mathbf{r}-\mathbf{R}_j)], \quad (26)$$

where V_c is the volume of the unit cell. Equation (26) gives the transition matrix for exciting a single electron from the *m*th state to the *n*th state with a momentum transfer $\mathbf{q}$.

The calculation of f^j_{nm} in general involves complicated quantum theory. However, under some reasonable approximation, the element f^j_{nm} can be derived analytically based on the hydrogen-like model for some light elements (Bethe, 1933; Madison & Merzbacher, 1975), such as carbon and oxygen *K*-shell ionizations.

4. Thermal diffuse scattering

Unlike single-electron excitation, thermal diffuse scattering (TDS) involves a large amount of excited states of different momentum transfers but the transition probability of each is very small. In this case, one neglects the inelastic transitions among all these excited states, and considers the fact that the TDS process does produce a large amount of momentum transfer but almost no energy loss; then (18) can be

simplified to

$$\varphi_0(\mathbf{b}, z) = \left\{ \exp(-i\sigma h'_{00}) \left[\varphi_0(\mathbf{b}, z_0) - i\sigma \sum_{n\neq 0} h'_{0n}\varphi_n(\mathbf{b}, z_0) \right] \right\} * P_0, \quad (27a)$$

$$\varphi_n(\mathbf{b}, z) = \{\exp(-i\sigma h'_{00})[\varphi_n(\mathbf{b}, z_0) - i\sigma h'_{n0}\varphi_0(\mathbf{b}, z_0)]\} * P_0. \quad (27b)$$

Since all the excited states have the same energy level, then the inelastic waves $\varphi_n s$ $(n = 1, 2, \ldots)$ have the same time-dependent factor $\exp(iEt/\hbar)$, and they can be considered approximately as coherent, because the different momentum transfer in TDS means that the electron is 'elastically' scattered off the beam direction. This may be similar to the electron elastic scattering off Bragg spots. By defining a total TDS wave φ^T as a coherent sum of all the TDS waves of different momentum transfers,

$$\varphi^T = \sum_n \varphi_n, \quad (28a)$$

and defining

$$h'_T = \sum_n h'_{n0}, \quad (28b)$$

from (27b) one has

$$\varphi^T(\mathbf{b}, z) = \{\exp(-i\sigma h'_{00})[\varphi^T(\mathbf{b}, z_0) - i\sigma h'_T\varphi_0(\mathbf{b}, z_0)]\} * P_0. \quad (29)$$

Now let us find the total generation function h'_T of a TDS wave. Based on quantum-mechanical theory, Takagi (1958) has given a perturbation potential of phonon scattering in a monoatomic crystal system,

$$U(\mathbf{r}) = -i\frac{V_c}{(2\pi)^3}\int \sum_j (\boldsymbol{\tau}\cdot\mathbf{u}_j) V_\tau \times \exp[-M(\tau)] \exp[i\boldsymbol{\tau}\cdot(\mathbf{r}-\mathbf{R}_j)]\, d\boldsymbol{\tau}, \quad (30a)$$

where

$$V_\tau = V_c^{-1}\int v(\mathbf{r}) \exp(-i\boldsymbol{\tau}\cdot\mathbf{r})\, d\mathbf{r}, \quad (30b)$$

$v(\mathbf{r})$ is the atomic potential and M is the Debye-Waller factor,

$$M(\tau) = \tfrac{1}{2}\langle(\boldsymbol{\tau}\cdot\mathbf{u}_j)^2\rangle, \quad (30c)$$

where the sum over j is taken over all the atoms in the crystal. The angular brackets in (30c) denote the time average of the corresponding quantity. The Fourier coefficient of potential V_τ is related to the structure factor $F(\tau)$ by

$$V_\tau = -\frac{\hbar^2}{2m_0}\frac{4\pi}{V_c}F(\tau). \quad (30d)$$

By expressing the lattice displacement $\mathbf{u}_j$ in normal modes, Whelan (1965b) and Rez *et al.* (1977) have given a transition matrix for creating a phonon of frequencies $\omega(\mathbf{q}, t)$ and polarization unit vectors $\mathbf{e}_{q,t}$ (where $t = 1, 2, 3$ denote the three perpendicular polarizations),

$$H'_T = \langle N(t)+1|U(\mathbf{r})|N(t)\rangle = \sum_{\mathbf{g}}\sum_{\mathbf{q}}\sum_t H_{\mathbf{g}}^{n0}\exp[i(\mathbf{g}-\mathbf{q})\cdot\mathbf{r}], \quad (31a)$$

where

$$H_{\mathbf{g}}^{n0} = -i(M_aN)^{-1/2}(\mathbf{g}-\mathbf{q})\cdot\mathbf{e}_{\mathbf{q},t}V_{\mathbf{g}-\mathbf{q}} \times \exp[-M(\mathbf{g}-\mathbf{q})]\{\hbar[N(t)+1]/2\omega(\mathbf{q}, t)\}^{1/2}. \quad (31b)$$

$N(t)$ is the occupation number of the state $|N(t)\rangle$, M_a is the atomic mass and N is the total number of atoms in the crystal. For the first-order approximation, one uses the Einstein model of assuming that each atom is vibrating independently with frequency ω, and also that the polarization of the phonon is independent of $\mathbf{q}$; then

$$N(t) = [\exp(\vartheta_E/T)-1]^{-1}, \quad (31c)$$

where $\vartheta_E = \hbar\omega/k_B$ is the Einstein temperature, T is the crystal temperature and k_B is the Boltzmann constant. Putting (31) into (24), one has

$$H'_T = -iA[V_c/(2\pi)^3]\sum_t\sum_{\mathbf{q}}\sum_{\mathbf{g}}\exp[i(\mathbf{g}-\mathbf{q})\cdot\mathbf{r}] \times (\mathbf{g}-\mathbf{q})_t V_{\mathbf{g}-\mathbf{q}}\exp[-M(\mathbf{g}-\mathbf{q})], \quad (32a)$$

and

$$A = \hbar(2k_B\vartheta_E M_aN)^{-1/2}\{[\exp(\vartheta_E/T)-1]^{-1}+1\}^{1/2}, \quad (32b)$$

where $\mathbf{q}$ is restricted to the first Brillouin zone. The sum over $\mathbf{q}$ in (32a) is on the two-dimensional array of states on the surface σ' for which energy and momentum are conserved (in practice the Ewald sphere). The calculation of (32a) is actually a function of incident-beam direction.

In the first-order approximation, the radius of the Ewald sphere can be taken as very large, so that the surface of the sphere is approximately a plane in the first Brillouin zone. If the incident beam direction is chosen as the z axis, then the double sum of

$$\sum_{\mathbf{q}}\sum_{g_x}\sum_{g_y}$$

is actually an integration in the whole of reciprocal space, for $\tau = (g_x - q_x, g_y - q_y)$ is parallel to the (x, y) plane. By selecting the three polarization directions as the (x, y, z) axes, then (32a) can be written as

$$H'_T \simeq -A[S_c/(2\pi)^2]\sum_t\frac{\partial}{\partial x_t}\sum_{g_z}\int\{\exp[i(\tau_x x+\tau_y y)] \times \exp(ig_z z)V_\tau(\tau_x, \tau_y, g_z)\exp(-M)\}\, d\tau_x\, d\tau_y \quad (33a)$$

where S_c is the surface area of the unit cell. Using the inverse Fourier transform of V_τ and through some algebra, one gets

$$H'_T = -A\sum_t \frac{\partial}{\partial x_t}\{V'(\mathbf{r})\}, \qquad (33b)$$

where V' is the temperature-modulated crystal potential. Integrate (33b) for z from z_0 to z; then put the result into (29) and consider the perturbation result of the TDS on the crystal potential H'_{00}, *e.g.* adding a Debye–Waller factor in the crystal potential (Cowley, 1975). The relationship between the total TDS wave before and after penetrating a crystal slice may then be given by

$$\varphi^T(\mathbf{b}, z) = \left(\exp[i\sigma V'(\mathbf{b}, \Delta z)]\left\{\varphi^T(\mathbf{b}, z_0) + i\sigma A\sum_t\left[\frac{\partial}{\partial x_t}V'(\mathbf{b}, \Delta z)\right]\varphi_0(\mathbf{b}, z_0)\right\}\right) * P_0, \qquad (34a)$$

where

$$V'(\mathbf{b}, \Delta z) = \int_{z_0}^{z} V'(\mathbf{b}, z)\,\mathrm{d}z. \qquad (34b)$$

For the elastic scattered wave, an absorption factor needs to be introduced in order to characterize the intensity decay. According to the absorption coefficient defined in (20b), one can approximately write (27a) as

$$\varphi_0(\mathbf{b}, z) = \left\{\exp[i\sigma V'(\mathbf{b}, \Delta z)]\varphi_0(\mathbf{b}, z_0)\exp\left(-\left[\sigma A \times\sum_t \frac{\partial}{\partial x_t}V'(\mathbf{b}, \Delta z)\right]^2 \Big/ 2\Delta z\right)\right\} * P_0. \qquad (35)$$

Equations (34) and (35) outline the main scheme of calculating TDS waves in a monatomic system. The A coefficient depends on the total number of atoms in the crystal, which is the result of conservation of the total energy. In practical calculations A can be replaced by a displacement constant of the atom from its equilibrium lattice point. For a complicated system consisting of several types of atoms, the term

$$A\sum_t\left[\frac{\partial}{\partial x_t}V'(\mathbf{r})\right]$$

in (34) and (35) may be generalized as

$$\sum_t\left[\frac{\partial}{\partial x_t}\sum_j A_j v'_j(\mathbf{r}-\mathbf{R}_j)\right], \qquad (36)$$

where $v'_j(\mathbf{r}-\mathbf{R}_j)$ is the atomic potential of atom j located at $\mathbf{R}_j$, including thermal effect, A_j is the constant of A defined in (32b) for the jth atom with Einstein temperature ϑ_{E_j}, which actually describes the displacement of the atoms from its equilibrium position depending on temperature.

As shown by Cowley (1988) based on the physical-optics approach, TDS can be considered as a time-dependent perturbation in multislice calculation; then the first-order expansion of the slice transmission function, $\exp(i\sigma V)$, for a small perturbation of ΔV due to atomic movement, will be the generation function of the TDS wave in the slice, which is proportional to the first-order differentiation of the potential V. His result is equivalent to (34).

5. Discussion and concluding remarks

The derivation of (18) was based on the following approximations: (1) small-angle scattering, *i.e.* $\alpha^2 \ll 1$; (2) small slice thickness, so that the variation of H'_{nm} in the region $(z - z_0)$ is slow; and (3) small energy loss, $\varepsilon_n \ll E_0$. Conditions (1) and (3) are usually satisfied in high-energy electron diffraction. Condition (2) gives a restriction on the selection of the slice thickness. This situation is similar to the slice-thickness selection in simulating high-resolution electron images. Good accuracy can be obtained by selecting slice thickness up to a few ångströms. Besides the above conditions, the crystal thickness is also limited. This is because the error arising from the calculation of the first slice will 'propagate' to the later slices, so that the final error is an accumulation of all errors arising from each slice. According to Ishizuka & Uyeda (1977), the slice thickness Z is restricted by $kZ\alpha^4 \ll 1$. This condition is often satisfied. The multislice method, in principle, neglects the back scattering of the electrons, which is usually very weak, especially for high-energy electrons.

The derivation of (18) assumed that the incident electron beam direction is along the z axis. For non-parallel incidence, (18) is also valid apart from the replacement of σ with $\sigma = (k/k_z)\sigma$ and propagation function P by $P' = (k_z/k)P$ (Ishizuka, 1982).

The derivation of (18) does not assume the three-dimensional periodicity of the crystal structure. This gives the possibility of introducing crystal defects in calculations. It is feasible, in principle, that multiple-scattering effects can be included in the calculations. The multislice method can be applied not only for calculating the transmission electron diffraction but also for the reflection electron diffraction in the geometry of glancing-angle incidence (Wang, 1989*a*, *b*).

We have thus given a general multislice solution of Yoshioka's coupling equations, which is capable of including the effects of non-periodic crystal structures and inelastic transitions among all the excited states in the calculations. The waves going into a crystal slice and those coming out of the slice are connected by an exponential form of the transition matrix; the different-order expansion of this matrix gives different orders of transitions. This general theory reduces to a simplified form if only one excited

state is involved. This form is identical to the theory proposed earlier (Wang, 1989*a*) for calculating energy-filtered inelastic images. In other words, we have established the equivalence of the early theory (Wang, 1989*a*) with quantum mechanics. It is proved that the generation of the inelastic wave can be considered as a mean excitation process, the probability of which is $\Delta z/\Lambda$ within a slice. This may indicate that the inelastic scattering of low energy loss is localized over a large range.

The multislice scheme of simulating the energy-filtered single electron excitation (*K* shells or *L* shells) and the total thermal diffuse scattering waves have been outlined. The theory reduces to the results of the physical-optics approach under some approximations. It is expected that these theories should provide an easier and accurate way of calculating inelastic scattering of high-energy electron diffraction in the geometries of both transmission and reflection electron microscopy. Detailed calculation results will be reported in a separate paper.

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APPENDIX *A*

Derivation of equation (6) from equation (5)

Let us first consider the *n*th inelastic component. By defining U as

$$U_n(\mathbf{r}) = \sum_m (2m_0/\hbar^2) H'_{nm}(\mathbf{r}) \Psi_m(\mathbf{r}), \qquad (A.1)$$

then we can write (4*a*) as

$$(\nabla^2 + k_n^2)\Psi_n(\mathbf{r}) = U_n(\mathbf{r}). \qquad (A.2)$$

The solution of (*A*.2) can be found through the Green-function method,

$$G(\mathbf{r}-\mathbf{r}', \mathbf{k}_n) = -\exp[i(k_n|\mathbf{r}-\mathbf{r}'|)]/(4\pi|\mathbf{r}-\mathbf{r}'|) \qquad (A.3)$$

which satisfies

$$(\nabla^2 + k_0^2) G(\mathbf{r}-\mathbf{r}', \mathbf{k}_n) = \delta(\mathbf{r}-\mathbf{r}'). \qquad (A.4)$$

Then the solution of (*A*.2) can be written as

$$\Psi_n(\mathbf{r}) = \exp[i\mathbf{k}_n \cdot \mathbf{r}] + \int G(\mathbf{r}-\mathbf{r}', \mathbf{k}_n) U_n(\mathbf{r}')\, d\mathbf{r}'. \qquad (A.5)$$

Replacing Ψ by $\varphi \exp(i\mathbf{k}_n \cdot \mathbf{r})$ and using (*A*.1), we find that (*A*.5) becomes

$$\varphi_n(\mathbf{r}) = 1 - \sum_m (2m_0/4\pi\hbar^2) \times \int F(\mathbf{r}-\mathbf{r}', \mathbf{k}_n) H'_{nm}(\mathbf{r}') \varphi_m(\mathbf{r}')\, d\mathbf{r}', \qquad (A.6)$$

with

$$F(\mathbf{r}-\mathbf{r}', \mathbf{k}_n) = \frac{\exp\{i[k_n|\mathbf{r}-\mathbf{r}'| - \mathbf{k}_n \cdot (\mathbf{r}-\mathbf{r}')]\}}{|\mathbf{r}-\mathbf{r}'|}. \qquad (A.7)$$

Equation (*A*.6) is the *n*th row of equation (6*a*).

APPENDIX *B*

Proof of equation (14)

Equation (14) is proved using the method of mathematical induction. First let us consider the solution for $L = 1$. From (13),

$$f_1^{(n)}(\mathbf{b}, z) = \iiint_{z'=z_0}^{z'=z} P_1(\mathbf{b}-\mathbf{b}', z-z') \times \sum_m H'_{1m}(\mathbf{b}', z') \varphi_m(\mathbf{b}_0, z_0) \times P_m(\mathbf{b}'-\mathbf{b}_0, z'-z_0)\, d\mathbf{b}_0\, d\mathbf{b}'\, dz'. \qquad (B.1)$$

By using the method of stationary phase for the integration of the propagation function P (Ishizuka & Uyeda, 1977), *i.e.* for a slow variation function V in the region $(z-z_0)$, one has

$$\int V(\mathbf{b}', z') P_n(\mathbf{b}-\mathbf{b}', z-z') P_n(\mathbf{b}'-\mathbf{b}_0, z'-z_0)\, d\mathbf{b}' \simeq V(\mathbf{b}_0, z') P_n(\mathbf{b}-\mathbf{b}_0, z-z_0). \qquad (B.2)$$

If we integrate over $\mathbf{b}'$ and assume the slow variation of H'_{nm} in the region $(z-z_0)$ as well as $k_1 = k_m$, (*B*.1) becomes

$$f_1^{(n)}(\mathbf{b}, z) = \int P_1(\mathbf{b}-\mathbf{b}_0, z-z_0) \times \sum_m h'_{nm}(\mathbf{b}_0, \Delta z) \varphi_m(\mathbf{b}_0, z_0)\, d\mathbf{b}_0. \qquad (B.3)$$

It is obvious that (*B*.3) is the second row of equation (14). If $f_L^{(n)}$ satisfies (14), then $f_{L+1}^{(n)}$ will be obtained from equation (13).

$$\begin{bmatrix} f_{L+1}^{(0)}(\mathbf{b}, z) \\ f_{L+1}^{(1)}(\mathbf{b}, z) \\ \vdots \\ f_{l+1}^{(m)}(\mathbf{b}, z) \end{bmatrix} = \int d\mathbf{b}' \int_{z'=z_0}^{z'=z} dz' \times \begin{bmatrix} P_0(\mathbf{b}-\mathbf{b}', z-z') & 0 & \cdots & 0 \\ 0 & P_1(\mathbf{b}-\mathbf{b}', z-z') & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \cdots & P_m(\mathbf{b}-\mathbf{b}', z-z') \end{bmatrix} \times \begin{bmatrix} H'_{00}(\mathbf{b}', z') & H'_{01}(\mathbf{b}', z') & \cdots & H'_{0m}(\mathbf{b}', z') \\ H'_{10}(\mathbf{b}', z) & H'_{11}(\mathbf{b}', z') & \cdots & H'_{1m}(\mathbf{b}', z') \\ \vdots & \vdots & \vdots & \vdots \\ H'_{m0}(\mathbf{b}', z') & H'_{m1}(\mathbf{b}', z') & \cdots & H'_{mm}(\mathbf{b}', z') \end{bmatrix} \times \frac{1}{L!} \int d\mathbf{b}_0 \times \begin{bmatrix} P_0(\mathbf{b}'-\mathbf{b}_0, z'-z_0) & 0 & \cdots & 0 \\ 0 & P_1(\mathbf{b}'-\mathbf{b}_0, z'-z_0) & \cdots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \cdots & P_m(\mathbf{b}'-\mathbf{b}_0, z'-z_0) \end{bmatrix} \times \begin{bmatrix} h'_{00}(\mathbf{b}_0, \Delta z') & h'_{01}(\mathbf{b}_0, \Delta z') & \cdots & h'_{0m}(\mathbf{b}_0, \Delta z') \\ h'_{10}(\mathbf{b}_0, \Delta z') & h'_{11}(\mathbf{b}_0, \Delta z') & \cdots & h'_{1m}(\mathbf{b}_0, \Delta z') \\ \vdots & \vdots & \vdots & \vdots \\ h'_{m0}(\mathbf{b}_0, \Delta z') & h'_{m1}(\mathbf{b}_0, \Delta z') & \cdots & h'_{mm}(\mathbf{b}_0, \Delta z') \end{bmatrix}^L \begin{bmatrix} \varphi_0(\mathbf{b}_0, z_0) \\ \varphi_1(\mathbf{b}_0, z_0) \\ \vdots \\ \varphi_m(\mathbf{b}_0, z_0) \end{bmatrix} \qquad (B.4)$$

where $\Delta z' = z' - z_0$. Multiplying out the first three matrices in (*B*.4), and integrating with respect to $\mathbf{b}'$ using (*B*.2), we find that (*B*.4) becomes

$$\begin{bmatrix} f^{(0)}_{L+1}(\mathbf{b}, z) \\ f^{(1)}_{L+1}(\mathbf{b}, z) \\ \vdots \\ f^{(m)}_{L+1}(\mathbf{b}, z) \end{bmatrix} = \frac{1}{L!} \int d\mathbf{b}_0 \int_{z'=z_0}^{z'=z} dz'$$

$$\times \begin{bmatrix} P_0(\mathbf{b}-\mathbf{b}_0, z-z_0) & 0 & \dots & 0 \\ 0 & P_1(\mathbf{b}-\mathbf{b}_0, z-z_0) & \dots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \dots & P_m(\mathbf{b}-\mathbf{b}_0, z-z_0) \end{bmatrix}$$

$$\times \begin{bmatrix} H'_{00}(\mathbf{b}_0, z') & H'_{01}(\mathbf{b}_0, z') & \dots & H'_{0m}(\mathbf{b}_0, z') \\ H'_{10}(\mathbf{b}_0, z') & H'_{11}(\mathbf{b}_0, z') & \dots & H'_{1m}(\mathbf{b}_0, z') \\ \vdots & \vdots & \vdots & \vdots \\ H'_{m0}(\mathbf{b}_0, z') & H'_{m1}(\mathbf{b}_0, z') & \dots & H'_{mm}(\mathbf{b}_0, z') \end{bmatrix}$$

$$\times \begin{bmatrix} h'_{00}(\mathbf{b}_0, \Delta z') & h'_{01}(\mathbf{b}_0, \Delta z') & \dots & h'_{0m}(\mathbf{b}_0, \Delta z') \\ h'_{10}(\mathbf{b}_0, \Delta z') & h'_{11}(\mathbf{b}_0, \Delta z') & \dots & h'_{1m}(\mathbf{b}_0, \Delta z') \\ \vdots & \vdots & \vdots & \vdots \\ h'_{m0}(\mathbf{b}_0, \Delta z') & h'_{m1}(\mathbf{b}_0, \Delta z') & \dots & h'_{mm}(\mathbf{b}_0, \Delta z') \end{bmatrix}^L \begin{bmatrix} \varphi_0(\mathbf{b}_0, z_0) \\ \varphi_1(\mathbf{b}_0, z_0) \\ \vdots \\ \varphi_m(\mathbf{b}_0, z_0) \end{bmatrix}. \quad (B.5)$$

Note that $h'_{nm} \simeq H'_{nm}(z'-z_0)$ for $\Delta z' \to 0$; then, integrating by parts in (*B*.5) for z', one obtains

$$\begin{bmatrix} f^{(0)}_{L+1}(\mathbf{b}, z) \\ f^{(1)}_{L+1}(\mathbf{b}, z) \\ \vdots \\ f^{(m)}_{L+1}(\mathbf{b}, z) \end{bmatrix}$$

$$= \frac{1}{(L+1)!} \int d\mathbf{b}_0 \begin{bmatrix} P_0(\mathbf{b}-\mathbf{b}_0, \Delta z) & 0 & \dots & 0 \\ 0 & P_1(\mathbf{b}-\mathbf{b}_0, \Delta z) & \dots & 0 \\ \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & \dots & P_m(\mathbf{b}-\mathbf{b}_0, \Delta z) \end{bmatrix}$$

$$\times \begin{bmatrix} h'_{00}(\mathbf{b}_0, \Delta z) & h'_{01}(\mathbf{b}_0, \Delta z) & \dots & h'_{0m}(\mathbf{b}_0, \Delta z) \\ h'_{10}(\mathbf{b}_0, \Delta z) & h'_{11}(\mathbf{b}_0, \Delta z) & \dots & h'_{1m}(\mathbf{b}_0, \Delta z) \\ \vdots & \vdots & \vdots & \vdots \\ h'_{m0}(\mathbf{b}_0, \Delta z) & h'_{m1}(\mathbf{b}_0, \Delta z) & \dots & h'_{mm}(\mathbf{b}_0, \Delta z) \end{bmatrix}^{L+1} \begin{bmatrix} \varphi_0(\mathbf{b}_0, z_0) \\ \varphi_1(\mathbf{b}_0, z_0) \\ \vdots \\ \varphi_m(\mathbf{b}_0, z_0) \end{bmatrix}. \quad (B.6)$$

It is necessary to point out that the result in (*B*.6) is only valid for Δz very small. With (*B*.3) and (*B*.6) the mathematical induction is completed.

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